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(19)

(54) SYNTHETIC PAPERS AND METHOD OF MAKING THE SAME

(71) We, MITSUBISHI PETRO-CHEMICAL COMPANY LIMITED, a joint stock company of Japan, located at 4, 2-Chome, Marunouchi, Chiyoda-Ku, Tokyo-To, Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates generally to a laminated structure made of a multiplicity of laminated films and more particularly to a synthetic paper and to a method of making it.

The term "synthetic paper" as used herein refers to a paper-like laminar structure made in the form of thin sheets or films of synthetic resinous material employed for various uses, such as writing and printing, as distinquished from natural cellulose paper.

Synthetic papers made of thermoplastic resin or papers coated with polymeric emulsions are known for use in writing, printing and the common uses to which natural cellulose paper is put. Moreover, it has been proposed to orientate thermoplastic film laminated structures under elevated temperature conditions in a transverse direction to form homogeneous oriented film products having excellent physical and optical properties, such as are shown in U.S. Patent No. 3,380,868. Polymeric film structures having a matte finish and a cellulose structure have also been proposed. The finish is produced in these films by a filler which roughens the surface upon stretching of the film and renders the film receptive to marking by such instruments as a pencil, crayon or ball point pen. Typical of these films is that disclosed in U.S. Patent No. 3,154,461. Laminates comprising layers of orientated films of thermoplastic materials in which at least one of the outermost layers of film contains a suitable inert additive have also been proposed. Laminates of this type are particularly useful as films which may be written on by a pencil or crayon. Laminates of this type are disclosed in U.S. Patent 3,515,626.

In the known orientated films composed of thermoplastic materials and laminates thereof, no attempt has been made to develop microvoids therein open to the surface of the film for better adherence of ink or printing materials thereon, nor a distribution of the microvoids providing a cellular, fibrous structure in a paper like film layer, nor a distribution of voids relative to the whole paper-like film which is such that the feel of cellulose paper of the same thickness is imparted thereto, the paper-like film having a specific gravity which is lower than that of pulp cellulose paper, an improved ink reception and an improved printability, opacity and strength relative to the known cellulose pulp paper.

This invention provides a synthetic paper or paper-like laminate structure, in the form of sheets of film which has a specific gravity lower than that of natural cellulose paper, an improved receptivity to ink, and an improved printability, opacity and strength.

The invention also provides a new and improved synthetic paper which does not have any fly ash when burned and has a low calorific content, thereby reducing pollution.

Another object of the invention is to provide a synthetic paper from which paper products such as writing paper, envelopes, bags, cups or wrappers can be constructed, and to provide a strong synthetic paper that has excellent folding endurance in that it may be folded and unfolded along a fold thousands of times without degradation and rupture.

A further object is to provide a synthetic paper that has greater tensile strength and burst strength than natural cellulose paper of the same thickness as well as known synthetic papers, and good waterproof qualities and dimensional stability when subjected to humidity.

An additional object is to provide a new and improved method f producing a new and improved synthetic paper.

The synthetic paper according to the invention comprises a laminated film structure having a base film layer and a paper-like film layer laminated thereto. The base film layer is a film made of thermoplastic resin which may have 0 to 20% by weight of a fine inorganic filler, having a particle size of 0.3—30 microns, dispersed in the resin and is in a biaxially orientated state. The paper-like film layer is likewise made of a thermoplastic resin having 0.5 to 65% by weight of a fine inorganic filler, having a particle size of 0.3—30 microns, dispersed substantially uniformly in the resin thereof and is in a uniaxially orientated state. Microvoids are formed on and in the paper-like film layer and dispersed therein so that open cells or microvoids open to a surface thereof free of the base film layer and elongated microvoids are distributed in the paper-like film layer with fibrous-like portions of the resin among the microvoids to define a cellular fibrous structure. The paper-like film layer may have a thickness of at least 10 microns.

The method according to the invention provides for admixing the constituents for making a base film layer principally constituting a thermoplastic resin which may or may not be provided with up to 20% by weight of a fine inorganic filler of particle size 0.3—30 microns. The composition is formed into a base film and uniaxially orientated in in a machine or longitudinal direction by longitudinally stretching the base film to at least 1—3 times its original length. A second thermoplastic film layer comprising therein 0.5 to 65% by weight of particles of a fine (0.3—30 micron particle size), inorganic filler substantially uniformly dispersed in the resin thereof is laminated onto the base film layer. The resulting laminated structure is uniaxially orientated in a transverse direction while in the heated state so that the base film layer is biaxially orientated and the second film layer is in a uniaxially orientated state. The stretching is at least 2.5 times the original dimension of the transverse portion of the laminated structure being stretched so that the stretching in conjunction with the particles of fine filler develops within this second film layer microvoids open to a surface thereof free from the base film and distributed in the resin to define therein a cellular, fibrous structure so that the second film becomes a paper-like film layer and the resulting laminate is a sheet of synthetic paper-like material or synthetic paper usable for writing or printing thereon with known means, and for making products of a type made from cellulose paper. The microvoids preferably constitute at least 10% of the total volume of the paper-like film layer and preferably should not exceed 65%.

The invention also provides apparatus for manufacturing a synthetic paper-like laminate comprising a first extruder means for extruding a base film layer made of a thermoplastic resin, a first heating means for heating the extruded thermoplastic resinous base film layer; a first stretching means for stretching the base film layer longitudinally at least 1.3 times its original longitudinal dimension to orientate the resin of the base film layer; a second extruder means for extruding a paper-like film layer; a laminating means for laminating the paper-like film layer on the base film layer to form a laminate; and a second stretching means for stretching the laminate transversely at least 2.5 times its transverse dimension to develop stresses in conjunction with said filler particles effective to develop microvoids in the paper-like film layer, which open to a surface thereof free of the base layer and microvoids internally thereof distributed with fibre-like portions of the resin among the microvoids interiorly of the paper-like layer to define therein a cellular, fibrous structure.

Other features and advantages of a paper-like laminated structure of synthetic paper according to the invention will be better understood in conjunction with the accompanying drawings, in which:

Fig. 1 is a cross-sectional view diagrammatically illustrating one example of synthetic paper according to the invention.

Fig. 2 is an elevation of an apparatus for manufacturing the synthetic paper in Fig. 1;

Fig. 3 is a plan view of a photomicrograph of a paper-like layer of synthetic paper according to the invention;

Fig. 4 is a longitudinal sectional view of a photomicrograph of the paper in Fig. 3;

Fig. 5 is a plan view of a photomicrograph of a printed sheet of plastic synthetic paper according to the prior art;

Fig. 6 is a plan view of a photomicrograph of a printed sheet of paper according to the invention;

Fig. 7 is a diagram or plot illustrating the percentage of voids as related to the stretching operation;

Fig. 8 is a perspective view of a bag made of synthetic paper according to the invention;

Fig. 9 is a perspective view of an envelope made of synthetic paper according to the invention;

Fig. 10 is a perspective view of a wrapper base made of a synthetic paper according to the invention; and

Fig. 11 is a plan view of an embossed wallpaper made of a synthetic paper according to the invention.

The synthetic paper according to the invention is constructed as a multiple-layer or multilayer sheet laminate 1, comprising a base film

layer 2 made of a thermoplastic resin.

A paper-like film layer 3 made of a thermoplastic resin mixed with particles of a fin inorganic filler within a range of 0.5 to 65% by weight is caused to adhere or is laminated to a surface of the layer 2. The synthetic paper may be made solely of the two layers 2 and 3, or may also include a second paper-like film layer 5 adhered to or laminated to the base film layer 2 on an opposite surface to that to which the first paper-like film layer 3 is adhered.

The base film layer 2 imparts the physical strength characteristics of the synthetic paper according to the invention as herein described. The paper-like film layers 3 and 5 provide improved surfaces on which writing and printing may be accomplished with the known writing and printing instruments as hereinafter described.

The base film layer 2 and the paper-like film layer 3 are formed from compositions which are later described herein in detail. The base film layer thermoplastic resin, with whatever additives have been added thereto, is formed by an extruder 7 which is of a single screw type having an L/D ratio of over 28:1. The film 2 is extruded through a film-forming die 8 having a uniformity index of over 0.9. The film 2 is delivered longitudinally over a set of rolls 10, 11 and passes through the nip of the rolls where its uniformity of thickness is maintained at a desired thickness as hereinafter disclosed. The film is delivered over guide rolls 12, 13 to a longitudinal stretching zone or machine 15 which uniaxially orientates the film in the longitudinal or machine direction. The film passes over a heating roll 15*a*, or it may be otherwise heated by infra-red heating devices, not shown. The heated thermoplastic film is stretched by two stretching rolls 15*b*, 15*c*. The film is stretched at least 1.3 times its longitudinal dimension to orientate the thermoplastic resin therein and to improve the physical strength characteristics of the film that will become the base layer film 2. The stretched film is cooled by a cooling roll 15*d*.

The uniaxially orientated base layer film is advanced longitudinally over guide rolls 17, 18 to a set of laminating rolls 20, 21 where it is laminated to a second thermoplastic film which will become the film layer 3 described hereinafter as to composition and characteristics. The layer 3 is extruded by a film-forming extruder 23 which is diagrammatically illustrated and is a vent-type extruder having and L/D ratio similar to that of the first described extruder and a die with a similar uniformity index. The laminating takes place in the nip of the laminating rolls 20, 21 at a pressure avoiding the formation of undulations in the surface of the films while maintaining pressure sufficient to obtain a strong bond between the two laminated film layers 2 and 3.

The double film or two-layer laminate is advanced longitudinally over guide rolls 25, 26, 27 and is advanced to a second set of laminating rolls 30, 31 near which is disposed a third extruder 33 of the same type as the second extruder 23 delivering a thermoplastic resin film, constituting the third film layer 5, laminated under pressure to a surface of the base film opposite to the first lamination of the first film thereon. The three-ply laminate 1 is advanced longitudinally over guide rolls 35, 36, 37 as illustrated where cooling takes place and is advanced to a second stretching apparatus 39 divided into three zones. A first zone is a pre-heating zone where the three-layer laminate is heated, for example, with hot air or infra-red heating devices to a temperature in the range of 140°C to 180°C and is then stretched in a transverse direction at least 2.5 times the original dimensions of the portions of the laminate between the stretching grippers of a stretching device in the second or stretching zone. The laminate is advanced from the stretching zone to a third zone or setting zone in which the laminate is reduced in temperature to from 120°C to 160°C. The temperature of the laminate in the stretching zone is from 140°C to 170°C.

The set laminate 1 may then be delivered to a take-up apparatus. However, a preferred embodiment of the method of the invention provides for a surface treatment to improve the adherence of ink to the two outermost film layers 3, 5 by polarising the two outermost films. We have found that the adherence of ink can be improved by applying to the surface of the outermost film layers a corona discharge treatment accomplished by electrodes 41, 42 on opposite sides of the laminate to which a voltage of 3000 to 30,000 volts is applied with a plate current of 0.5 to 5 amperes. Treatment is accomplished by maintaining the clearance between the laminate and the electrodes 41, 42 substantially constant. Accordingly, stabilizing rolls 44, 45 maintain the film laminate 1 and the electrode clearance accurately, it being understood that the electrodes are disposed immediately adjacent to the stabilising rolls downstream thereof and extend transversely of the film laminate 1.

The invention provides for a composition of the outermost layers of the laminate to provide paper-like layers on which writing and printing can take place with known means. The composition of the paper-like layers is described hereinafter. However, the composition of the thermoplastic resin with the controlled filler content and minimum size of filler particles constituting 0.3 micron and the uniform distribution of the filler provides a composition in which microvoids can be controllably developed by the transverse stretching of the laminate. The microvoid formation is a function of the size of filler particles, the extent of stretching and the filler content which is

from 0.5% to 65% by weight, preferably at least 20% by weight, and more particularly at least 25% by weight f the whole of the composition from which the paper-like layers 3 and 5 are formed.

The stretching of the laminate 1 must be sufficient to develop an internal mechanism within the outermost films during the stretching in which inorganic filler particles 50 in each of the outermost film layers 3 and 5 coact with the stretching to develop microvoids 51 in the film layers. The transverse stretching must be at least 2.5 times the original dimension as before described. Stretching of seven times the dimension and even more for example, up to sixteen times, has yielded suitable voids. Thus, as the stretching takes place, stresses develop about the uniformly dispersed filler particles 50, and microvoids or cells 51 are developed. Some cells 52 are formed next adjacent to the surface of the outermost films 3 and 5 free of the base film layer 2. These cells or voids 52 are open to the surface and in communication therewith as illustrated in Fig. 3. The transverse orientation of the paper-like film layers 3, 5 results in the formation of microvoids 51 internally of the film structure as illustrated in Fig. 4. The distribution of the internal microvoids 51 is such that portions 55 of the resin among the microvoids are fibrous in structure as clearly shown in Fig. 4. The internal cells or microvoids 51 may be independent, and some will communicate with others. Generally, each microvoid 51 has internally thereof a filler particle 50 effecting the formation of a microvoid. The filler particles generally are in contact with two opposite sides or boundaries 50a, 50b of the resin within which the microvoid is formed as is clearly seen in Fig. 4. The microvoids 52 on the surface of the paper-like film layers 3, 5 provide the synthetic paper with a better receptiveness to ink than other synthetic paper or film. The surface likewise has the "feel" of natural cellulose papers, and the surface is improved for writing thereon with instruments such as pencils, crayons or ball point pens.

The microvoids 51 formed interiorly of the outermost film layers reduce the density and specific gravity of these layers so that the resulting synthetic paper 1 is much lighter than known synthetic writing films and papers including coated natural cellulose papers and art paper. The fibrous, cellular structure of the outer films 3, 5 provides a cusionability improving the printing and ink transfer during printing with the known techniques. Accordingly the ink transfer to the surface on the printed areas is substantially uniform as illustrated in Fig. 6 as compared with known plastic coated paper as illustrated in Fig. 5 and other synthetic films and papers.

The microvoid formation and particularly the surface microvoids improve the light reflective characteristics of the synthetic paper according to the invention so that the whiteness and opacity are better than those of natural cellulose papers and synthetic films and papers. The improved whiteness and opacity improve the ability to reproduce the desired printed image.

These improved characteristics in conjunction with the improved cusionability and ink receptiveness result in the improved printing and the quality thereof mentioned heretofore. The ink dots 67 of the printing on a printed paper 69 according to the invention (Fig. 6) are distributed uniformly on the surface, and spaces or gaps of the type shown at 70, 71 on a known coated paper in Fig. 5 are avoided. The uniformity of the printing assists in the reproduction of the desired image.

The reduced density and cellular structure throughout the synthetic paper 1 improve its anti-pollution characteristic in that there is a reduction in solid matter and the voids provide spaces and areas readily accessible to air so the paper will burn more readily with less release of heat and less calorific value. The synthetic paper 1 will burn without smoke and fly ash in that complete combustion takes place and the ashes remain in the container within which the burning takes place.

The base film layer 2 is principally a thermoplastic resin, polyolefin resins such as homopolymers and copolymers of, for example, ethylene, propylene and butene-1, polyamide resins, polyester resins such as polyethylene terephthalate, polyvinyl resins such as homopolymers and copolymers of vinyl chloride and polyvinylidene resins such as homopolymers and copolymers of vinylidene chloride and homopolymers and copolymers of styrene can be used singly or as mixtures thereof. Moreover, auxiliary materials such as stabilizers, plasticisers, fillers and pigments may be contained in the base film layer material resin within its stretching range as necessary. When less than 20% by weight of a fine inorganic filler is blended into the base layer film resin, some favourable results can be obtained. These results are an improved whiteness, opacity, stiffness and toughness of the resulting synthetic paper. Moreover, the surface of the base film layer 2 becomes coarse, and adhesion of the other two films thereto is greatly improved.

An orientatable thermoplastic resin is used for the paper-like film layers 3 and 5 and may be the same or different resin chosen from the group of the above-mentioned resins used in the base film layer. When a heat sealing characteristic is desired, the softening point of the paper-like film layer must be less than that of the base film resin. For example, in the event that the base film layer is made of a homopolymer, and the paper-like film layer is made of a copolymer containing a monomer which is a component unit of the homopolymer, in the base film layer the resulting

paper-like film layer will have a lower softening point than that of the base layer. Under these conditions, a favourable heat sealing characteristic can be developed in the paper and heat sealing can be carried out free from shrinkage of the base film layer.

The choice of the resins must be such that the resins in the paper-like film layers have a sufficient adhesiveness with respect to the base film layer so that lamination can be carried out effectively. The use of a filler in the base film layer develops microvoids on the surface thereof improving the adhesion of the paper-like layers thereto. Moreover, an anchor coat can be applied to the base film layer to improve the adhesion of the paper-like film layers, for example using a polyisocyanate or a polytitanate. The resin for the paper-like film layer may also contain suitable auxiliary materials as in the case of the base layer resin.

The paper-like film layer resin contains particles of a fine inorganic filler as listed hereinafter. Care must be taken in choosing the filler that it should be a filler which does not discolour due to decomposition at elevated temperatures which are applied in the course of making the synthetic paper according to the invention. Examples of suitable inorganic fillers for the paper-like film layers are: clay, talc, asbestos, gypsum, barium sulphate, calcium carbonate, magnesium carbonate, magnesium oxide, diatomaceous earth, kieselguhr, titanium dioxide, zinc oxide, silica, a silicate or silicon oxide particles used either singly or as a mixture of two or more of these fillers. The filler must be a fine filler of a grain size 0.3 to 30 microns, preferably 0.5—30 microns. The paper-like film layers contain 0.5% to 65% and preferably 5% to 60% by weight of a fine inorganic filler in the component materials.

As heretofore explained, the addition of the inorganic filler in the formation of microvoids and accordingly the lightness, stiffness and toughness of the paper-like are greatly improved. The printability, whiteness and the feel of the overall paper are improved, because of the generation of voids. Moreover, the surface of the or each paper-like film layer may be oxidised to give a greater ink attraction. The paper-like layer may also be given a greater ink attraction by applying an electrostatic charge to the synthetic laminate.

The resin of the paper-like film layers is prepared for delivery to the film-forming extruders 23, 33 by taking 100 parts of the resin and separating it into two batches. A batch of 90 parts of the resin has filler added thereto, for example in an amount of 20 parts. A smaller master batch of resin is prepared which constitutes 10 parts of the original 100 parts of the resin to which are added an antistatic agent, the stabilizer as hereinafter described and a dispersing agent in small amounts for example, 0.3 to 1 percent by weight. The dispersing agent assists in obtaining a uniform distribution of the inorganic filler particles. The two separate batches are independently mixed in a Henshel mixer and after mixing of the batches the two batches are then mixed together again in a Henshel mixer. The resulting mixture of the two batches is then delivered to a twin-screw extruder in order to improve the uniformity of the mixture and the uniformity of distribution of the inorganic filler throughout the composition. The output from the twin screw mixer is the starting material or product pellets which are delivered to the film-forming extruders 23, 33 heretofore described with respect to Fig. 2.

The composition used for making the base film layer is treated similarly to the preliminary steps defined heretofore with respect to the mixing and extrusion of the paper-like film layers. However, in the event that only a small amount of filler be used for example, 3% to 5% by weight, the materials are mixed in a single batch in a Henshel mixer and then delivered to a twin screw extruder where they are pelletised and the starting material for the film-forming extruder 7 is thus made.

The relation or ratio between stretching and void formation is illustrated in the graph in Fig. 7. The graph illustrates curves indicating that the stretching and the filler content control the ratio of voids to the whole of the individual paper-like film layers. The range of the curves illustrate that transverse stretching of at least 2.5 times (X 2.5 as shown) will form voids, and that the stretching can exceed 15.0 times (X 15) the original transverse dimention.

The following lists and tables set forth examples of the components used in preparing the several film layers and the ratios and manufacturing conditions:

The term "machine direction" means a longitudinal direction, and the "stretching ratio" is the ratio of the length of film after stretching to the length before stretching.

Resins:	Polypropylene
PP	Polyethylene
PE	Ethylene-vinyl acetate polymer
EVA	Ethylene-propylene-copolymer
EPP	Polyvinyl chloride
PVC	Polyvinylidene chloride
PVDC	Polystyrene
PS	High impact polystyrene
HI/PS	Acrylonitrile butadiene styrene
ABS	copolymer

Fillers:

Kieselguhr, clay, Ca-carbonate, TiO_2, Barium sulphate, Magnesium oxide.

Stabilisers:

Calcium stearate
Aluminium stearate
Zinc stearate
Calcium laurylate
Titanium dioxide
Barium stearate
Topanol (Trade Mark) 1,1,3 - tris - (2-methyl - 4 - hydroxy - 5 - *t* - butylbenzyl) - butane

Anti-static agents:

Polyoxyalkylamine

NYMEEN-S-210 (made by Nippon Oils and Fat Co. Ltd.)

$$C_{18}H_{37}N<\begin{matrix}(CH_2CH_2O)_xH\\(CH_2CH_2O)_yH\end{matrix}$$

Tertiary alkylamine; $x+y=10$; Molecular wt. 709; Vis. 500 cps.

Aliphatic glycerine ester

RESISTAT-PE-132 (made by Daiichi Kogyo Seiyaku Co. Ltd.) monoglyceride of $C_{17}H_{35}COOH$

$$\begin{matrix}CH_2OCOC_{17}H_{35}\\|\\CHOH\\|\\CH_2OH\end{matrix}$$

molecular weight 358 RESISTAT-PEO139 almost the same kind as above.

Alkyl phosphate

PHOSPHANOL-SM-1 (made by Toho Chemical Industries Co. Ltd.)

$$P\begin{matrix}\diagup(CH_2CH_2O)_nOR\\—(CH_2CH_2O)_nOR\\\diagdown(CH_2CH_2O)_nOR\end{matrix}$$

R=alkyl with carbon number 18.

PHOSPHANOL-NP-10. The same substance as above except R=alkyl with carbon number 12.

Alkyl Betaine

LEOSTAT-532 (made by Lion Fat and Oil Co. Ltd.)

$$R—N^{\oplus}\begin{matrix}\diagup(CH_2CH_2O)_nH\\—(CH_2CH_2O)_nH\\\diagdown CH_2COOH^{\ominus}\end{matrix}$$

Dispersion Agents:

Polyoxylene alkyl ether

EMULGEN (made by KAO-ATLAS Co. Ltd.)

$C_{18}H_{37}O(CH_2CH_2O)_nH$ ($n=10$ Molecular weight 710)

Alkyl phosphate ester

OM-11 (made by Marubishi Petrochemical Co. Ltd.) $(RO)_2P(O)(OX)$ R=alkyl with carbon number 12—18. X=amine, of viscosity 500 cps, boiling point 330°C.

Polyoxyalkylamine

NYMEEN-T-210 (made by Nippon Oils and Fats Co. Ltd.)

$$R—N<\begin{matrix}(CH_2CH_2O)_xH\\(CH_2CH_2O)_yH\end{matrix}$$

$x+y=10$

R=unsaturated alkyl with carbon number approximately 10. R: tallow alkyl

Anchor coating agents:

Polyisocyanate

EL-250 (made by Toyo Ink Manufacturing Co. Ltd.) polyisocyanate that has R—NCO monomeric units, where R is a hydrocarbon group.

Polytitanate

EL-110 (made by Toyo Ink Manufacturing Co. Ltd.)

Examples 5, 6 and 8 in Table II (a), Examples 10 to 18 in Table II (b), Example 8 in Table VII (a) and Example 17 in Table VII (b) are comparison Examples.

TABLE I (a)

COMPONENTS: BASE LAYER

Ex. No.	Resin	Parts	Filler	Parts	Stabilizer	Parts	Anti-static Agent	Parts	Dispersing Agent	Parts	Other Ingredients	Parts
1	PP	100	Clay Kieselguhr	5 10	Ca-stearate Topanol	0.3 0.1	PHOSPHANOL SM-1 (Toho-Chem.Inc)	0.1	NYMEEN S-210 (Nippon Oil and Fat Co. Ltd)	0.1		-
2	PP PE	90 10	Clay	5	Ca-stearate Topanol	1.0 0.1	PHOSPHANOL SM-1 (Toho Chem. Inc)	0.3	NYMEEN S-210 (Nippon Oil and Fat Co. Ltd)	0.3		-
3	PP Polybutene-1 PE	80 10 10	Clay	25	Al-stearate Topanol	1.0 0.5	LEOSTAT-532 (Lion Fat & Oil Co.)	0.5	EMULGEN (Kao Atlas Co.)	0.6		-
4	PP EVA (VA 12%)	80 20	Clay	3	Zn-stearate Topanol	0.5 0.1	LEOSTAT-532 (Lion Fat & Oil Co.)	1.0	EMULGEN (Kao Atlas Co.)	0.5	Ultraviolet proofing agent	0.3
5	PE EVA (VA 15%)	80 20	-		Ca-stearate Topanol	1.0 0.1	-		-			-
6	PP EPP (E 3%)	90 10	-		Ca-laurate Topanol	0.5 0.5	-		EMULGEN (Kao Atlas Co.)	0.5		-
7	PP EPP (E 15%)	90 10	Clay Ca-carbonate	2 10	Zn-stearate Topanol	0.5 0.3	-		EMULGEN (Kao Atlas Co.)	0.3		-
8	PP EPP (E 98%)	90 10	Clay TiO_2	15 10	Ca-stearate Topanol	1.3 0.7	-		EMULGEN (Kao Atlas Co.)	0.3		-
9	PE	100	Clay	10	Topanol	0.1	-		EMULGEN (Kao Atlas Co.)	0.3		-
10	PVC PVDC	80 20	barium sulphate clay	3 2	Ba-stearate	1.5	-		EMULGEN (Kao Atlas Co.)	1.0		-

TABLE I (b)

COMPONENTS: BASE LAYER

Ex. No.	Resin	Parts	Filler	Parts	Stabilizer	Parts	Anti-static Agent	Parts	Dispersing Agent	Parts	Other Ingredients	Parts
11	PP PE-terephthalate	80 20	Clay	1	-		-		NYMEEN S-210 (Nippon Oil & Fat Co)	0.1	-	
12	PP Polyamide (Nylon-6)	80 20	Kieselguhr	0.5	-		-		NYMEEN S-210 (Nippon Oil & Fat Co)	0.1	-	
13	PP	100	-		Ca-stearate	0.5	-		-		-	
14	PP	100	-		Ca-stearate	0.5	-		-		-	
15	PS	100	-		-		-		-		-	
16	PE	100	-		Ca-stearate	0.5	-		-		-	
17	PE HI/PS	80 20	Clay Kieselguhr	1 1	Ca-stearate Topanol	0.5 0.3	NYMEEN S-210 (Nippon Oil & Fat Co.)	0.5	- -		- -	
18	PE ABS	80 20	Clay Kieselguhr	2 7	Al-stearate Topanol	1.0 0.3	RESISTAT PE 132 (Daiichi Kogyo Seiyaku)	0.5	-		-	

Natural Cellulose Paper - weight percent of filler or other components can be calculated by the following formula:

$$\text{weight percent of filler} = \frac{\text{Filler Parts}}{\text{Total Parts of Resin, Filler and Additives}} \times 100$$

TABLE II (a)

COMPONENTS: PAPER-LIKE LAYER

Ex. No.	Resin	Parts	Filler	Parts	Stabilizer	Parts	Anti-static Agent	Parts	Dispersing Agent	Parts	Others	Parts
1	PP	100	Clay Kieselguhr	15 10	Ca-stearate Topanol	0.5 0.3	PHOSPHANOL SM-1 (Toho Chemical Inc.)	0.1	NYMEEN S-210 (Nippon Oil & Fat Co.)	0.7	-	
2	PP PE	80 20	Clay	5	Ca-stearate Topanol	1.0 0.1	PHOSPHANOL SM-1 (Toho Chem. Inc.)	0.3	NYMEEN S-210 (Nippon Oil & Fat Co.)	1.0	-	
3	PP Polybutene-1 EVA(VA 10%)	80 10 10	Clay	40	Al-stearate Topanol	0.5 0.5	PHOSPHANOL SM-1 (Toho Chem. Inc.)	0.5	NYMEEN S-210 (Nippon Oil & Fat Co.)	1.0	-	
4	PP EVA(VA12%)	80 20	Clay Kieselguhr	23 10	Zn-stearate Topanol	0.5 1.0	NYMEEN S-210 (Nippon Oil & Fat Co.)	0.3	NYMEEN S-210 (Nippon Oil & Fat Co.)	0.3	Ultraviolet proofing agent	0.3
5	PVC	100	Clay Ca-carbonate	50 80	Ca-stearate Ba-stearate	1.0 1.0	NYMEEN S-210 (Nippon Oil & Fat Co.)	1.0	NYMEEN S-210 (Nippon Oil & Fat Co.)	0.5	-	
6	PP EPP(E 3%)	90 10	Clay Kieselguhr TiO_2	70 40 20	Ca-laurate Topanol	0.5 0.5	NYMEEN S-210 (Nippon Oil & Fat Co.)	0.5	EMULGEN (Kao Atlas Co.)	0.5	-	
7	PP EPP(E 15%)	30 70	Clay Ca-carbonate	10 15	Zn-stearate Topanol	0.3 0.3	NYMEEN S-210 (Nippon Oil & Fat Co.)	0.5	EMULGEN (Kao Atlas Co.)	0.5	-	
8	PE EPP(F 15%)	80 20	Clay Kieselguhr	100 50	Topanol	0.2	-		-		-	
9	EPP(E 20%)	100	Clay	33	Topanol	0.1	-		-		-	

TABLE II (b)

COMPONENTS: PAPER-LIKE LAYER

Ex. No.	Resin	Parts	Filler	Parts	Stabilizer	Parts	Anti-static Agent	Parts	Dispersing Agent	Parts	Others	Parts
10	PVC	100	Clay	100	Ba-stearate Ca-stearate	1.5 0.5	-		-		-	
11	PP PE-terephthalate	80 20	Clay	50	-		RESISTAT PE 132 (Daiichi Kogyo Seiyaku Co.)	0.3	NYMEEN S-210 (Nippon Oil & Fat Co.)	0.1	-	
12	PP Polyamide (Nylon-6)	80 20	Clay	80	-		"	0.7	"	0.5	-	
13	PP Polyamide (Nylon-6)	80 20	Clay Magnesium Oxide	80 20	-		"	0.7	"	0.5	-	
14	PP PS	80 20	Clay Ca-carbonate	70 30	-		"	0.7	"	0.5	-	
15	PS	100	Clay TiO_2	90 10	-		"	0.7	"	0.5	-	
16	PE PS	80 20	Clay Barium sulphate	70 10	-		"	0.7	"	0.5	-	
17	PE HI/PS	80 20	Clay Kieselguhr	80 30	Ca-stearate Topanol	0.7 0.5	NYMEEN S-210 (Nippon Oil & Fat Co.)	0.5	-		-	
18	PE ABS	80 20	Clay Ca-carbonate	20 15	Ca-stearate Topanol	0.7 0.5	NYMEEN S-210 (Nippon Oil & Fat Co.)	0.5	-		-	

TABLE III

MANUFACTURING CONDITIONS

Ex.	BASE LAYER: Stretching Temperature °C	BASE LAYER: MD (Machine Direction) stretching ratio (x) (times initial length)	PAPER-LIKE LAYER: Temperature: laminating C°	PAPER-LIKE LAYER: Temperature: preheating C°	PAPER-LIKE LAYER: Temperature: Stretching C°	PAPER-LIKE LAYER: Temperature: Setting C°	PAPER-LIKE LAYER: Stretching ratio (x)
1	150	5	260	160	160	140	8
2	140	2	300	160	150	140	5
3	140	4	250	160	150	130	7
4	140	6	250	150	145	130	6
5	130	1.3	130	150	140	120	4
6	140	5	280	165	145	140	5
7	140	5	280	160	150	140	8
8	150	4	280	160	150	140	7
9	135	3	250	160	150	140	2.5
10	100	5	130	110	100	85	6
11	145	4	280	165	160	140	5
12	145	5	250	165	160	140	7

TABLE III (continued)

MANUFACTURING CONDITIONS

Ex.	BASE LAYER: Stretching Temperature °C	BASE LAYER: MD (Machine Direction) stretching ratio (x) (times initial length)	PAPER-LIKE LAYER: Temperature: laminating C°	PAPER-LIKE LAYER: Temperature: preheating C°	PAPER-LIKE LAYER: Temperature: Stretching C°	PAPER-LIKE LAYER: Temperature: Setting, C°	PAPER-LIKE LAYER: Stretching ratio (x)
13	145	5	250	160	150	140	8
14	140	6	250	160	150	140	7
15	100	4	200	120	110	90	6.5
16	135	6	220	140	130	130	5
17	135	7	250	140	130	130	8
18	135	5	250	140	130	130	4

TABLE IV

THICKNESS OF ARTICLE

Ex.	BASE LAYER (microns) μ	PAPER-LIKE LAYER (microns) μ
1.	40	40
2.	50	60
3.	30	10
4.	20	10
5.	30	30
6.	60	20
7.	70	40
8.	30	50
9.	80	60
10.	50	20
11.	20	100
12.	40	50
13.	20	20
14.	20	10
15.	80	70
16.	40	80
17.	40	100
18.	50	30

TABLE V

PROPERTIES

Ex.	OPACITY (%)	TENSILE STRENGTH MD/TD (kg)	SPECIFIC GRAVITY	INK TRANSFER (g/m^2)	INK ADHESION	TEAR STRENGTH MD/TD (kg/cm)	FOLDING ENDURANCE (cycles)
1.	55	9.6/17.6	0.78	2.1	△	70/60	>150.000
2.	38	12.5/24.5	0.85	1.2	△	108/98	>150.000
3.	38	5.6/9.6	0.81	3.0	0	65/58	>150.000
4.	35	3.7/7.3	0.75	3.0	0	46/43	>150.000
5.	60	7.2/14.4	0.68	3.2	0	66/59	>150.000
6.	89	9.6/17.2	0.72	3.2	0	108/100	>150.000
7.	57	13.0/26.2	0.77	2.1	△	120/110	>150.000
8.	90	13.1/25.6	0.71	3.4	0	70/62	>150.000
9.	91	10.1/23.1	0.78	3.1	0	120/105	>150.000
10.	88	8.4/19.2	0.72	3.2	0	105/95	>150.000
11.	93	14.2/25.0	0.72	3.1	0	25/22	>150.000
12.	92	10.2/20.0	0.73	3.1	0	50/45	>150.000
13.	48	5.8/9.6	0.69	3.0	0	21/19	>150.000
14.	40	4.2/8.2	0.73	3.1	0	20/18	>150.000
15.	92	7.2/15.6	0.81	2.9	0	85/70	>150.000
16.	90	11.5/23.2	0.78	2.8	0	60/52	>150.000
17.	94	10.5/21.3	0.79	3.1	0	50/40	>150.000
18.	87	9.7/17.6	0.85	2.9	0	70/50	>150.000
Natural Cellulose Paper							
	85∿90	8.9/5.4	0.8∿1.2	2.3∿3.5	0	28/26	1000

Legend

MD – Machine Direction
TD – Transverse Direction
∿ – symbol for "approximately"
0 – good
△ – fair

TABLE VI

FILLER EFFECT IN BASE LAYER*

Filler Content (wt. %)	Stretching Stability (without splitting)	Tear Strength (after stretching)	Opacity	Whiteness
0	good	good	opacity	whiteness
10	good	good	increases as	increases as
20	good	good	filler	filler
25	inferior (film split)	inferior	increases	increases
30	poor (film split)	poor	↓ good	↓ good

* Resin = PP

Filler = Kaolinite clay

TABLE VII (a)

EFFECT OF FILLER CONTENT IN PAPER-LIKE LAYER

		Properties - A				Properties - B		
Ex. (Resin= PP)	Filler Content (wt.%)	Hardening time of ink (min)	Ink adhesion	Ink transfer (g/m^2)	Opacity (%)	Gloss (%)	Surface intensity (IGT cm/sec)	Smoothness (Beck sec)
1	0.5	1000	△~X	0.8	7	60	360	3000
2	5	820	△	1.2	38	45	360	1700
3	20	240	0	2.8	88	36	290	1200
4	25	200	0	2.9	89	35	280	1150
5	30	180	0	3.0	90	34	270	1120
6	60	95	0	3.4	94	22	200	800
7	65	80	0	3.5	95	20	130	500
8	70	70	0	3.5	95	15	55	100
Natural Cellulose Paper		50~250	0	2.5~3.5	85~90	3~40	100~200	100~1000

Properties A = Improve as filler content increases
Properties B = Decrease as filler content increases
IGT cm/sec = Test method used
Beck sec = The higher in value indicates improved smoothness
△ = Fair
X = Inferior
0 = Good
Resin = PP
Filler = Kaolinite clay

TABLE VII (b)

EFFECT OF FILLER CONTENT IN PAPER-LIKE LAYER ON VOID FORMATION

				Properties - A				Properties B		
Ex.	Void proportion* in paper-like layer (%)	Filler content (wt %)	Stretching extent (x times)	Hardening time of ink (min)	Ink adhesion	Ink transfer (g/m^2)	Opacity (%)	Gloss (%)	Surface intensity (IGT cm/sec)	Smoothness (BECK cm/se)
9	5	5	8	820	△	1.2	38	45	360	1700
10	10	20	4	300	△	2.1	57	42	360	1500
11	15	20	5	250	0	2.8	88	38	300	1250
12	20	25	5	200	0	2.9	89	36	290	1200
13	30	35	8	170	0	3.0	92	34	270	1120
14	59	60	12	95	0	3.4	94	24	210	1000
15	60	63	9	90	0	3.4	95	22	200	800
16	65	65	15	80	0	3.5	95	20	130	500
17	70	70	15	70	0	3.5	95	15	55	100
Natural Cellulose Paper			50∿270		0	2.3∿3.5	85∿90	3∿40	100∿200	100∿1000

* Void Proportion of Paper-like layer is as follows:

$$V\ (\%) = \frac{Po - P}{Po} \times 100$$

where V: Void Proportion
Po: Initial Density of paper-like layer
P: Density of paper-like layer after stretching

△ = fair
0 = Good
Resin= PP
Filler ; Kaolinite clay and Kieselguhr where ratio = $\frac{KC}{K} = \frac{4}{1}$

In the tables above, if the tables with a numeral and a letter are placed one above the other in proper sequence of the examples listed therein and in a descending order from left to right, an overall view of the components, manufacturing conditi ns and resulting characteristics of the manufactured papers will be readily seen.

The tables VII (a) and VII (b) indicate the effect of filler on various properties of the synthetic paper and provide a comparison with natural cellulose paper. As can be seen from the tables, the synthetic paper according to the invention has improved qualities over paper made from natural pulp.

The synthetic papers according to the invention are suitable for use as high grade printing paper for cards, maps, charts, decorative papers, dull finished or mat papers, typing, duplicating, drafting, tracing, writing, drawing, packaging, wrapping, paper boards and the like.

In articles manufactured from the synthetic paper of the invention, the synthetic paper may comprise sheet material with one part thereof overlying another part thereof and bonded thereto. The two parts may be integrally fused together as a weld along at least a portion in which the parts are in registry with and overlying one another.

The synthetic paper made according to the invention provides a paper with sufficient toughness and strength to be used for making bags as shown in Fig. 8 in which a shopping bag 70 is made out of a single sheet of synthetic paper. The sheet is folded in a conventional manner and made into the bag. The longitudinal seam along the length of the bag and folds forming the bottom of the bag are bonded by the use of an adhesive or bonding agent. Bonding can be effected by heat seals or "welds". When heat seals or bonds are effected, suitable pressures, times and temperatures and current values are used as is well known in this art. The bag is provided with handles 75, 76 made, for example, of synthetic filaments anchored to the bag by strips 77, 78 of rigid material, for example, paper or cardboard, underlying folds 79, 80 at the mouth of the bag.

Not only is the synthetic paper very strong and adequate for this use but the exterior of the bags may be decorated more clearly with coloured printing pictures as compared with the case when conventional printing paper is used. Thus, the pictures such as those of a city or mountain scenes or aeroplanes may be reproduced as desired. The possibility of using heat for forming bonds makes it possible to manufacture envelopes 81 as shown in Fig. 9 in which all of the cutting, folding and bonding at the seams 82, 83, 84 are accomplished automatically by heat sealing. Moreover, the bonding or seaming may be accomplished by using vibrations to effect the bonds in making this type of product. The envelope can be decorated as desired.

In effecting heat seals electrically heated bars are used, for example. The heat seals are effected in tw to three seconds at a pressure of substantially 2 kg/cm² and at a temperature of from 200°C to 220°C. The current pulses are applied to the bars at a rate of from 0.2 second to one second with a current value of from twenty to thirty amperes. The strength of the heat seals increases from one second of the application of heat to two seconds, and a longer application of heat does not materially improve the strength of the seal.

Ultrasonic heat sealing is accomplished in two to three seconds when the vibratory cycle is 28 Kc. Furthermore, heat sealing by using induction or resistance heating of the sealing elements is possible.

The synthetic paper can be used as packaging and wrapping paper. Fig. 10 illustrates a roll 90 of synthetic paper 91 made according to the invention for wrapping an item 92. Those skilled in the art will understand that the item 92 can be a box or package to be wrapped or can be an article of food, or any article or articles such as a stack of cards or papers to be wrapped, for example automatically. The synthetic paper 91 can likewise be decorated as desired.

As indicated heretofore, a particular advantage of the paper-like laminate according to the invention is that it is waterproof and can be folded and unfolded thousands of times without failure or rupture. This feature makes it possible to make maps and charts having a long life when subjected to moisture and folding and unfolding. The waterproof characteristics of the synthetic paper of the invention improve its use as a wall covering.

The synthetic paper can be used to make wallpaper 95 illustrated in Fig. 11. The wallpaper may be embossed as illustrated at 96 or may have any desired decorations, including printing, thereon.

Those skilled in the art will understand that laminates formed (18 examples) according to Tables I(a) to Table IV have properties as set forth in Table V which are generally superior to those of natural cellulose paper. Some effects of a filler on a PP base layer can be seen from Table VI. It can be seen from the latter table that, in order to maintain good mechanical strength in the base layer, the filler should preferably not exceed 20% by weight of the base layer composition.

The properties of the PP compositions (17 examples) with filler therein as noted are set forth in Tables VII(a) and VII (b) and the properties designated A and B are generally superior to natural cellulose paper.

Those skilled in the art will recognise that microvoid formation is essential to the paper-like layer of the present invention and these

microvoids are elongated and formed in dependence upon the extent of stretching and the size of the particles of the filler whose diameter principally determines the major width dimensions of the voids. The number of microvoids developed is principally controlled by the percentage by weight of filler in the paper-like layer compositions. The length of the voids is essentially controlled by the extent of the stretching and this is designated (x) in the various tables.

Thus, the paper-like layers of the various laminates disclosed are fibrous internally and are able to have images printed thereon and accept various inks, and can be written on by various instruments.

The synthetic paper according to the invention has excellent tear and burst strength and the heat seals or bonding of seams likewise are of great strength. Hot wet adhesives or any type of suitable adhesive can likewise be used to make suitable bonds.

It can be seen that the principal constituents in the paper are the resins and the filler components. The stabilizers are only from 0.1% to 2% by weight; the antistatic agents are 1.0% to 1.5% by weight; the latter value is true for the dispersion agents and the ultraviolet light-proofing agents. The synthetic paper according to the invention can be made without the additional components, those other than the principal components, listed above.

WHAT WE CLAIM IS:—

1. A synthetic paper comprising: (a) a base film layer comprising a thermoplastic resin, the base film layer being in a biaxially orientated state; and (b) a paper-like film layer laminated to the base film layer and composed of a thermoplastic resin having from 0.5% to 65% by weight of particles of a fine, inorganic filler, having a particle size of 0.3 to 30 microns, dispersed substantially uniformly in the resin thereof, the paper-like film layer being in a uniaxially orientated state and having microvoids dispersed therein, including microvoids open to a surface thereof free of the base film layer (a), and the microvoids comprising elongated microvoids distributed in the paper-like film layer (b) with fibrous-like portions of the resin among the microvoids interiorly of the paper-like film layer (b) to define therein a cellular, fibrous structure.

2. A synthetic paper according to claim 1, in which substantially each microvoid contains a particle of the inorganic filler.

3. A synthetic paper according to claim 1, in which the base film layer (a) comprises up to 20% by weight of a fine inorganic filler, having a particle size of 0.3—30 microns, dispersed in the resin thereof, and in which the filler content of the paper-like film layer (b) is substantially greater than that of the base film layer (a).

4. A synthetic paper according to any of claims 1 to 3 in which the filler comprises particles of 0.5 to 30 microns in size.

5. A synthetic paper according to any of claims 1 to 4, in which the filler in the paper-like film layer comprises from 20% to 65% by weight of the total layer (b).

6. A synthetic paper according to claim 5, in which the filler comprises from 25% to 60% by weight of the total layer (b).

7. A synthetic paper according to any of claims 1 to 6 including a second paper-like film layer laminated to the base film layer on a surface thereof opposed to that to which the first-mentioned paper-like film layer is laminated, the second paper-like film layer comprising a thermoplastic resin having 0.5 to 65% by weight of a fine, inorganic filler, of particle size 0.3 to 30 microns, dispersed substantially uniformly in the resin thereof, and the second paper-like film layer being in a uniaxially oriented state and having microvoids dispersed therein including microvoids open to a surface free of the base film layer, and the microvoids, comprising elongated microvoids distributed in the second paper-like film layer with fibrous-like portions of the resin among the microvoids interiorly of the paper-like film layer defining therein a cellular, fibrous structure.

8. A synthetic paper according to any of claims 1 to 7, in which the paper-like film layer comprises said fibre-like portions of resin extending generaly in a same direction.

9. A synthetic paper according to claim 1, in which the proportion of microvoids in the paper-like film layer is from 10% to 65%.

10. A synthetic paper according to any of claims 1 to 9 in which the base film layer comprises an antistatic agent.

11. A synthetic paper according to any of claims 1 to 10, in which the or each paper-like film layer comprises an antistatic agent.

12. A synthetic paper according to any of claims 1 to 11, in which the base film layer comprises a stabiliser.

13. A synthetic paper according to any of claims 1 to 12 in which the or each paper-like film layer comprises a stabiliser.

14. A synthetic paper according to any of claims 1 to 13, in which the base film layer comprises an ultraviolet light-proofing agent.

15. A synthetic paper according to claim 1, in which the or each paper-like film layer comprises an ultraviolet light-proofing agent.

16. A synthetic paper according to any of claims 1 to 15, in which the inorganic filler comprises fine particles of clay, talc, asbestos, gypsum, barium sulphate, calcium carbonate, magnesium carbonate, kieselguhr, titanium dioxide zinc oxide, magnesium oxide, diatomaceous earth, silica, a silicate or silicon oxide, or any mixture thereof.

17. A synthetic paper according to any of claims 1 to 16, in which the resin of the base

film layer, or the paper-like film layer, or of either paper-like film layer is a polyolefin resin such as a homopolymer or copolymer of ethylene, propylene or butene-1, a polyamide resin, a polyester resin such as polyethylene terephthalate, a polyvinyl resin such as a homopolymer or copolymer of vinyl chloride, a polyvinylidene resin such as a homopolymer or copolymer of vinylidene chloride, or a homopolymer or copolymer of styrene.

18. A synthetic paper according to claim 17, in which the base film layer and the paper-like film layer are composed of identical resins.

19. A synthetic paper according to claim 17, in which the base film layer and the or each paper-like film layer comprise different resins chosen from the group of resins listed in claim 17.

20. A synthetic paper according to claim 17 or 19 in which the resin in the base film layer and the resin in the or each paper-like film layer comprises a polypropylene.

21. A synthetic paper according to any of claims 1 to 20 in which the base film layer comprises microvoids which are fewer in number than the microvoids in the or each paper-like film layer.

22. A synthetic paper according to claim 1, in which the surface of the or each paper-like film layer is oxidised to give a greater ink attraction.

23. A method of producing a synthetic paper-like laminate comprising providing a base film layer composed of a thermoplastic resin, uniaxially orientating the base film layer in a machine or longitudinal direction, by longitudinally stretching it to at least 1.3 times its original length, laminating on the base film layer a paper-like film layer composed of a thermoplastic resin having 0.5 to 65 % by weight of particles of a fine inorganic filler, of particle size 0.3—30 microns, substantially uniformly distributed therein so as to form a laminate, stretching while in a heated state the laminate in a transverse direction so as to biaxially orientate the base film layer and uniaxially orientate the paper-like film layer, including stretching the laminate in the transverse direction at least 2.5 times more than the original dimension of the laminated films in the transverse direction to develop microvoids in the paper-like film layer in conjunction with the particles of filler distributed in the paper-like layer and to develop fibrous-like portions of the resin thereof among the microvoids interiorly of the paper-like film layer so as to define therein a cellular, fibrous structure.

24. A method according to claim 23, including applying an electrostatic charge to the synthetic laminate in order to give the paper-like layer a greater ink attraction.

25. A method according to claim 23 or 24 in which the base film layer is extruded, immediately afterwards cooled and stretched while in heated state in a longitudinal direction so as to orientate the resin thereof, and in which the laminate is stretched transversely.

26. A method according to claim 23, in which the laminate is preheated and stretched transversely so as biaxially to orientate the base film layer and uniaxially to orientate the paper-like film layer.

27. A sheet of printed synthetic paper comprising a base film layer comprising a thermoplastic resin, the base film layer being in a biaxially orientated state, a paper-like film layer laminated to the base film layer and composed of a thermoplastic resin having from 0.5% to 65% by weight of particles of a fine, inorganic filler, of particle size 0.3 to 30 microns, dispersed in the resin thereof substantially uniformly, the paper-like film layer being in a uniaxially orientated state and having microvoids dispersed therein including microvoids open to a surface thereof free of the base film layer, and the microvoids comprising elongated microvoids distributed in the paper-like film layer with fibrous-like portions of the resin among the microvoids interiorly of the paper-like film layer to define therein a cellular, fibrous structure, and a plurality of ink dots closely spaced and distributed uniformly throughout an area on the surface of the paper-like film layer defining thereon an image.

28. An article of manufacture comprising a synthetic paper, said synthetic paper comprising a base film layer comprising a thermoplastic resin, the base film layer being in a biaxially orientated state, a paper-like film layer laminated to the base film layer and composed of a thermoplastic resin having from 0.5% to 65% by weight of particles of a fine, inorganic filler, of particle size 0.3—30 microns, dispersed substantially uniformly in the resin thereof, the paper-like film layer being in an uniaxially orientated state and having microvoids dispersed therein including microvoids open to a surface thereof free of the base film layer, and the microvoids comprising elongated microvoids distributed in the paper-like film layer with fibrous-like portions of the resin among the microvoids interiorly of the paper-like film layer to define therein a cellular, fibrous structure.

29. An article of manufacture according to claim 28, comprising synthetic sheet material folded and bonded in the configuration of a container.

30. An article of manufacture according to claim 29, in which said container comprises a bag, and handles on said bag.

31. An article of manufacture according to claim 29 or 30 in which said resin in said synthetic paper comprises a resin rendering said synthetic paper waterproof.

32. An article of manufacture according to any of claims 29 to 31 in which said container

has the configuration of an envelope.

33. An article of manufacture according to claim 28 in which said synthetic paper comprises a roll of wallpaper.

34. An article of manufacture according to claim 28, in which synthetic paper comprises a wrapper on another article of manufacture.

35. An article of manufacture according to claim 28, in which said synthetic paper comprises sheet material with one part thereof overlying another part thereof and bonded thereto.

36. An article of manufacture according to claim 35, in which said one part and said another part are integrally fused together as a weld along at least a portion in which said parts are in registry with and overlying one another.

37. Apparatus for manufacturing a synthetic paper-like laminate according to any of claims 1 to 22 comprising a first extruder means for extruding a base film layer made of a thermoplastic resin; a first heating means for heating the extruded thermoplastic resinous base film layer; a first stretching means for stretching the base film layer longitudinally at least 1.3 times its original longitudinal dimension to orientate the resin of the base film layer; a second extruder means for extruding a paper-like film layer; a laminating means for laminating the paper-like film layer on the base film layer to form a laminate; and a second stretching means for stretching the laminate transversely at least 2.5 times its transverse dimension to develop stresses in conjunction with said filler particles effective to develop microvoids in the paper-like film layer, which open to a surface thereof free of the base layer and microvoids internally thereof distributed with fibre-like portions of the resin among the microvoids interiorly of the paper-like layer to define therein a cellular, fibrous structure.

38. A synthetic paper comprising a base film layer comprising a thermoplastic resin, the base film layer being in a biaxially orientated state, a paper-like film layer laminated to the base film layer and made of a thermoplastic resin having from 0.5% to 65% by weight of particles of a fine, inorganic filler having a particle size of 0.3 to 30 microns dispersed substantially uniformly in the resin thereof, the paper-like film layer being in a uniaxially orientated state and having microvoids dispersed therein including microvoids open to a surface thereof free of the base film layer, and the microvoids comprising elongated microvoids distributed in the paper-like film layer with fibre-like portions of the resin among the microvoids interiorly of the paper-like film layer to define therein a cellular, fibrous structure, and the resin being a resin receptive to ink and to writing thereon.

39. A synthetic paper according to claim 38, in which said paper-like film layer has a thickness of at least 10 microns.

40. A synthetic paper according to claim 1 substantially as herein described with reference to the specific examples.

41. A method according to claim 23 substantially as herein described with reference to the accompanying drawings and/or any of the specific examples.

42. A sheet of printed paper according to claim 27 substantially as herein described with reference to the accompanying drawing and/or any of the specific examples.

43. An article of manufacture according to claim 28 substantially as herein described with reference to the accompanying drawings and/or any of the specific examples.

44. Apparatus according to claim 37 substantially as herein described with reference to the accompanying drawings and/or any of the specific examples.

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1384556 COMPLETE SPECIFICATION

5 SHEETS *This drawing is a reproduction of the Original on a reduced scale*

Sheet 1

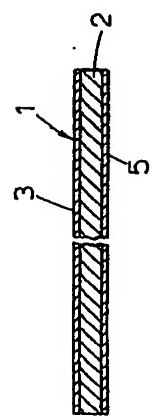


Fig. 1.

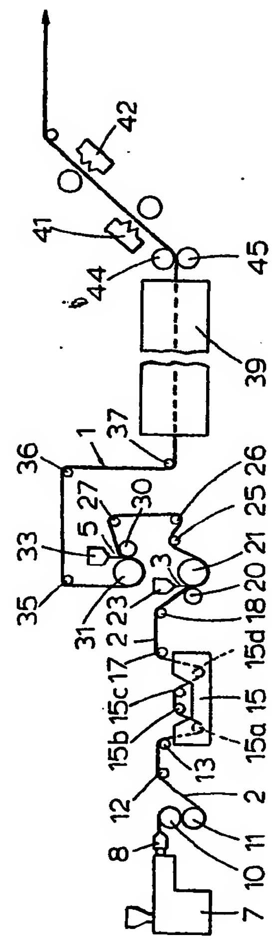


Fig. 2.

Fig. 3.

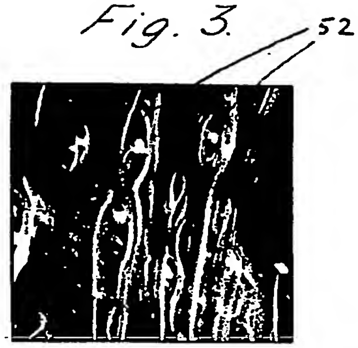


Fig. 4.

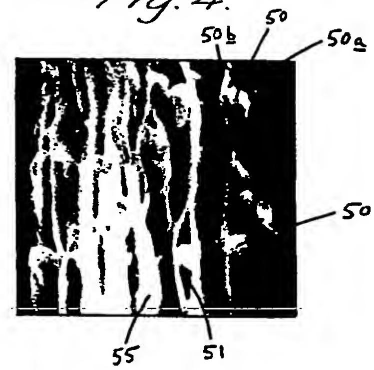


Fig. 5.

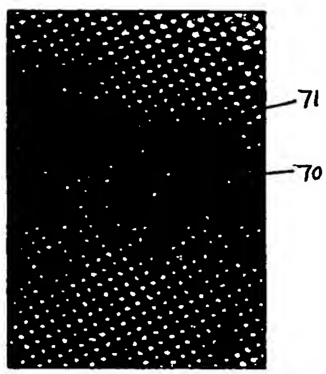
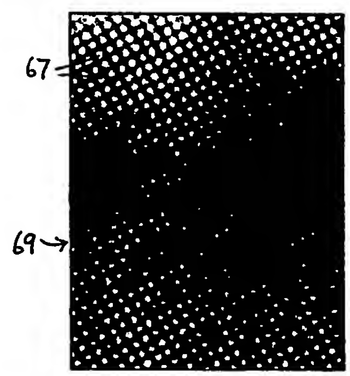


Fig. 6.



VOID PROPORTION VS FILLER CONTENT
(PAPER-LIKE LAYER)

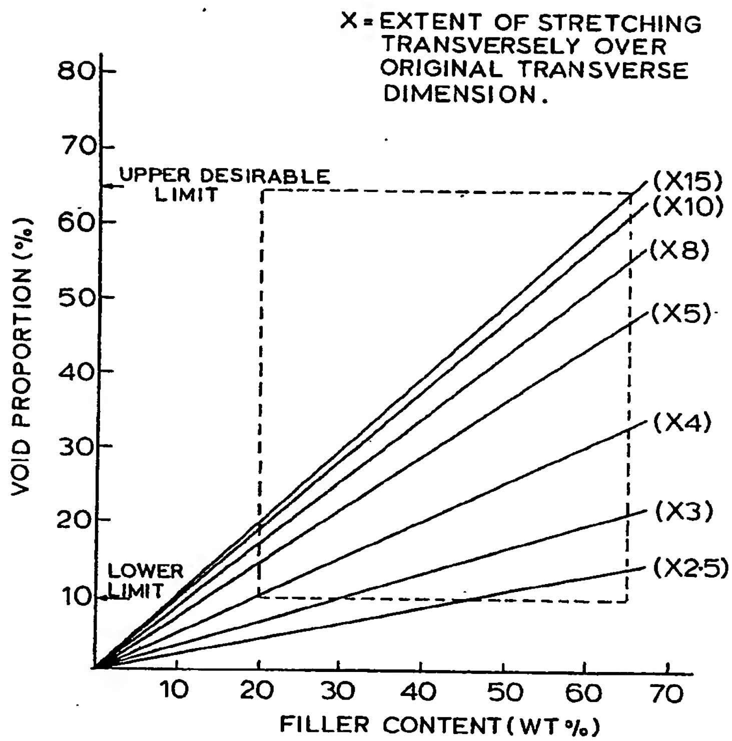


Fig.7.

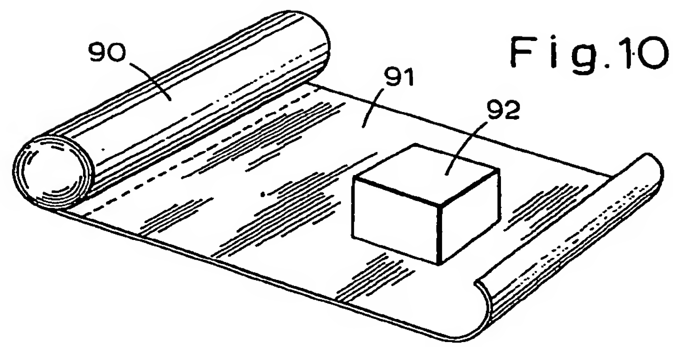


Fig.10.

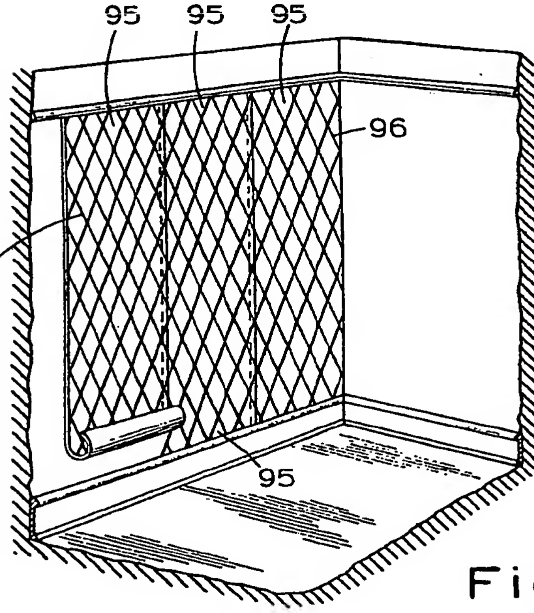


Fig.11.

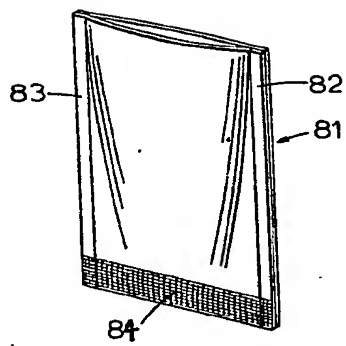

83
82
81
84

Fig. 9.

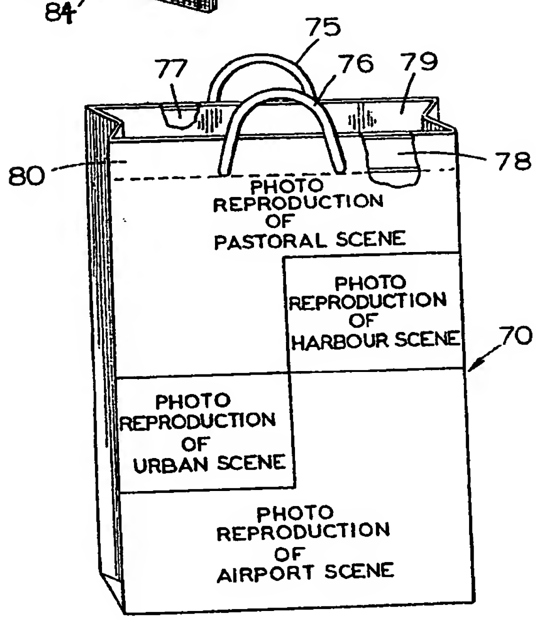

75
77
76
79
80
78
PHOTO
REPRODUCTION
OF
PASTORAL SCENE
PHOTO
REPRODUCTION
OF
HARBOUR SCENE
70
PHOTO
REPRODUCTION
OF
URBAN SCENE
PHOTO
REPRODUCTION
OF
AIRPORT SCENE

Fig. 8.